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Final Report

NASA Origins of Solar System Program: Grant NAGW 3065 "Molecular Cloud Diagnostics of Solar Nebula Chemistry"

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The purpose of this proposal was to measure two diagnostic abundance ratios, N2/NH3 and CH4/CO, in a statistical sample of galactic star-forming regions using mm-wave astronomy, and compare them with ratios found in comets in order to constrain models of solar nebula chemistry. Part of this proposal concerned laboratory millimeter-wave spectroscopy of two isotopically substituted species, CH3D and <sup>14</sup>N<sup>15</sup>N.

During the past 3 years of our grant, we have achieved a large fraction of the goals. We have estimated N<sub>2</sub> abundances from observations of N<sub>2</sub>H<sup>+</sup> towards a large sample of molecular clouds. From these measurements, we were able to derive N<sub>2</sub>/NH<sub>3</sub> ratios for all of these objects, which were subsequently compared with ratio values for comets. We also carried out observations of CH<sub>3</sub>D towards several molecular clouds, from which we obtained upper limits to the interstellar methane abundance. Towards one cloud, Orion-KL, the limits were significant enough to estimate the CH<sub>4</sub>/CO ratio, which was then compared with cometary values. We also carried out laboratory measurements for CH<sub>3</sub>D, which were used in our astronomical studies. Furthermore, we have a tentative measurement of the pure rotational spectrum of <sup>14</sup>N<sup>15</sup>N. The details of our results are described in the following sections.

A) N<sub>2</sub>/NH<sub>3</sub> Ratio

Establishing this ratio in the interstellar medium was primarily done during Year 1 of this proposal. During this period, the interstellar N2 abundance was established through observations of its protonated form, N2H+, which has several pure rotational transitions at millimeter wavelengths. (N2 cannot be observed directly in cooler gas because it lacks a permanent dipole moment.) From these measurements, N<sub>2</sub> abundances were inferred. These works were published in 1992 (Womack, Ziurys, and Wyckoff, Ap. J., 387, 417; Womack, Ziurys, and Wyckoff, Ap. J., 393, 188). Our N2 abundances were then compared with those for ammonia found in the literature. These values are the first N2/NH3 ratios published to date, and typically it was found that  $N_2/NH_3 \sim 170$ ; in comparison, the  $N_2/NH_3$  ratio observed in comet Halley is N2/NH3 ~ 4. Hence, considering gas components only, the average molecular cloud value for this ratio is ~40 times larger than that found in the comet. Consequently, the abundances of these nitrogen gases in comet Halley do not reflect those found in dense molecular clouds, strongly suggesting that the comet nucleus appears to be comprised of material that has undergone chemical processing that likely occurred in the early solar nebula (Womack, Wyckoff and Ziurys, Ap. J., 401, 728).

These conclusions about the N2/NH3 ratio are based on interstellar N2 abundances inferred from N2H<sup>+</sup>. A direct measurement of the N2 abundance would be preferable, which could be done by studying its singly 15-substituted isotopomer  $^{14}N^{15}N$ , which has a small, but non-zero, dipole moment. The rotational rest frequencies of  $^{14}N^{15}N$  are not yet available but they would provide a more direct measurement of the interstellar molecular nitrogen abundance. During Year 2 of this proposal, we attempted to measure the millimeter-wave transitions of  $^{14}N^{15}N$  near 230 and 345 GHz. We believe we have a tentative detection of the J = 2  $\rightarrow$  1 and J = 3  $\rightarrow$  2 transitions at 230 and 345 GHz (see Fig. 1), but need a third, confirming line at 460 GHz. This measurement involved purchasing a frequency quadrupler device for the spectrometer system, which did not work properly at first and had to be returned to the manufacturer, Millitech Corp. Furthermore, it was found that a lower loss feedhorn had to be fabricated, which caused additional delay.

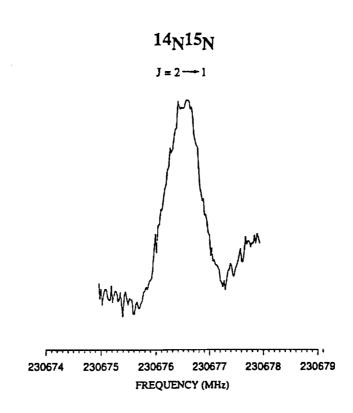


Fig. 1. Tentative laboratory spectrum of  $^{14}N^{15}N$ :  $J = 2 \rightarrow 1$ .

During Year 3, the equipment for the 460 GHz measurements was together and working, but then the sample cell kept breaking. However, we have solved the

problems with the sample cell and hope to complete the 460 GHz measurements soon. After we have done this work, we will search for this species in dense interstellar clouds using radio telescopes and consequently obtain N<sub>2</sub> abundances by direct observation of the molecule. N<sub>2</sub> has not yet been detected in any astronomical source. We will use these observations to refine the current interstellar N<sub>2</sub>/NH<sub>3</sub> abundance ratios. Comparison of these ratios with existing chemical models of the early solar nebula and dense cloud ion-molecule chemistry will provide the first direct observational constraints for the chemical evolution of planets and comets.

## B) CH<sub>4</sub>CO Ratio

While CO is ubiquitous in dense clouds with a well-determined abundance, the situation is different for CH<sub>4</sub> because it does not have a permanent dipole moment and hence is not readily observable in the cooler material present in most molecular gas. The singly deuterated form of methane, CH<sub>3</sub>D, however, has rotational transitions which, in principle, can be observed in dense clouds. Over the past three years, we have attempted to detect CH<sub>3</sub>D in such objects. This work also included new laboratory measurements of CH<sub>3</sub>D, done in Year 2. The first two years of work resulted in what appeared to be a tentative detection of CH<sub>3</sub>D in three dense molecular clouds: Orion-KL, SgrB2N, and W51M. We observed one possible line of CH<sub>3</sub>D at 232 GHz and two possible transitions at 465 GHz, using primarily the Cal Tech Sub-mm Observatory (CSO).

The 465 GHz lines needed additional integration to substantiate their presence, and hence the detection of interstellar CH<sub>3</sub>D. In Year 3, we had three observing runs at the CSO to carry out these confirming measurements. The first two sessions were unsuccessful because of poor weather conditions. In the third and final run, the weather was excellent and outstanding signal-to-noise was obtained at 465 GHz in Orion-KL. Unfortunately, while one line was present at the CH<sub>3</sub>D frequencies, the other one clearly was not. Hence, CH<sub>3</sub>D was not detected down to significant levels of  $T_A*<0.05$  K in Orion-KL. These observations suggest an upper limit to the CH<sub>3</sub>D column density of  $6\times10^{18}$  cm<sup>-2</sup> in the Orion "hot core" region, and a fractional abundance limit, relative to H<sub>2</sub>, of  $f(CH_4/H_2) < 6\times10^{-6}$ .

These measurements do place a constraint on the methane abundance in the Orion-KL hot core region. They indicate that the fractional abundance of CH<sub>4</sub> in this gas has a limit of  $< 6 \times 10^{-4}$ . This value in turn suggests CH<sub>4</sub>/CO  $\lesssim 6$  in Orion-KL. In contrast, a ratio of CH<sub>4</sub>/CO  $\sim 0.03$ -0.2 was found in comet Halley. This difference in the CH<sub>4</sub>/CO abundance ratio between the comet and star forming region suggests that significant chemical processing occurred in the outer solar nebula prior to comet formation, as also indicated by N<sub>2</sub>/NH<sub>3</sub> ratios. The chemical

significance of these contrasting ratios is analyzed in more detail by Wyckoff, Ziurys and Kleine (1995, Ap. J., submitted).

## Other Results

We also began to explore other abundance ratios in interstellar clouds vs. comets. In Year 2 we obtained some methanol and formaldehyde spectra, which we observed towards comet Swift-Tuttle at millimeter wavelengths using the NRAO 12 m telescope and the CSO. We are currently comparing the CH<sub>3</sub>OH/H<sub>2</sub>CO ratio in this comet to that found in molecular clouds (Womack et al., in preparation).

In addition, our survey of N<sub>2</sub>H<sup>+</sup> in molecular clouds yielded some interesting results for star formation. Towards Orion, our original survey spectra suggested evidence of two clouds which appear to be in physical contact near the region of KL/IRc2, i.e. where young stars are present. We subsequently followed up these data with observations at higher spatial resolution, using the IRAM 30 m telescope. These measurements confirmed the presence of at least two clouds, overlapping in the region of protostars. This data suggests that cloud/cloud interactions may help to trigger star formation (Womack, Ziurys, and Sage 1993 Ap. J., 406, L29).

## **Publications**

- "N<sub>2</sub>H+ in Orion: Chemical Clues to the Dynamics of the Quiescent Gas,"
  M. Womack, L. M. Ziurys, and S. Wyckoff, Ap. J. (Letters), 370, L99 (1991).
- 2. "A Survey of N<sub>2</sub>H<sup>+</sup> in Dense Clouds: Implications for Interstellar Nitrogen and Ion-Molecule Chemistry," M. Womack, L. M. Ziurys, and S. Wyckoff, Ap. J., 387, 417 (1992).
- 3. "Estimates of N<sub>2</sub> Abundances in Dense Molecular Clouds," M. Womack, L. M. Ziurys, and S. Wyckoff, Ap. J., 393, 188 (1992).
- "Observational Constraints on Solar Nebula Nitrogen Chemistry: N<sub>2</sub>/NH<sub>3</sub>,"
  M. Womack, S. Wyckoff and L. M. Ziurys, Ap. J., 401, 728 (1992).
- 5. "N<sub>2</sub>H+ in the Orion Ambient Ridge: Cloud Clumping vs. Rotation," M. P. Womack, L. M. Ziurys and L. J. Sage, Ap. J. (Letters), 406, L29 (1993).
- 6. "A Search for Interstellar CH<sub>3</sub>D: Limits to the Methane Abundance in Orion-KL," M. P. Womack, A. J. Apponi, and L. M. Ziurys, 1995 *Ap. J.*, submitted.

7. "Do Comet Volatiles Represent Unprocessed Interstellar Gases?" S. Wyckoff, L. M. Ziurys, and M. Kleine, 1995, Ap. J., submitted.

## **Manuscripts in Preparation**

1. "Comparison of Methanol and Formaldehyde Abundances in Comet Swift-Tuttle," M. Womack, F. P. Schloerb, L. M. Ziurys, D. Saunders, and J. Deane, in preparation.